Spatio-temporal variations in aerosol properties over western India

A THESIS

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by

Rajesh T. A.

Under the Supervision of

Prof. S. Ramachandran
Guide
Space and Atmospheric Sciences Division
Physical Research Laboratory
Ahmedabad, India

Prof. P. C. Vinodkumar
Co-Guide
Department of Physics
Sardar Patel University
Vallabh Vidyanagar, India

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Date: 17/Sep/2018

CERTIFICATE

Certified that the work incorporated in this thesis entitled **"Spatio-temporal variations in aerosol properties over western India"**, submitted by **Mr. Rajesh T. A.** comprises the results of independent and original investigations carried out. The materials obtained from other sources and used in the thesis have been duly acknowledged appropriately.

Place: Vallabh Vidyanagar
Date: 17/Sep/2018
Registration No.: 640, Date: 29/10/2013

Rajesh

Rajesh T. A.
(Research Student)

Certified that the work mentioned above was carried out under my/our guidance.

Prof. S. Ramachandran
(Research Guide)

Prof. P. C. Vinodkumar
(Research Co-Guide)

Forwarded Through:

Prof. P. Janardhan
Dean
Physical Research Laboratory
Ahmedabad

प्रो. जनार्दन पद्मनाभन
Prof. Janardhan Padmanabhan
डीन, भौतिक अनुसंधान प्रयोगशाला
Dean, Physical Research Laboratory
(भारत सरकार, अंतरिक्ष विभाग की यूनिट)
(Unit of Department of Space, Govt. of India)
नवरंगपुरा, अहमदाबाद-380 009 भारत
Navrangpura, Ahmedabad-380 009 India

Prof. P. C. Vinodkumar
Head
Department of Physics
Sardar Patel University

HEAD
DEPARTMENT OF PHYSICS
SARDAR PATEL UNIVERSITY
VALLABH VIDYANAGAR

DECLARATION

*I, Rajesh T. A., S/o Mr. T. K. Ayyappen, resident of D-5, PRL Residences, Vikramnagar, Bopal-Ambli Road, Ahmedabad - 380058, hereby declare that the research work incorporated in the present thesis entitled "***Spatio-temporal variations in aerosol properties over western India***" is my own work and is original. This work (in part or in full) has not been submitted to any University for the award of a Degree or a Diploma. I have properly acknowledged the material collected from secondary sources wherever required. I solely own the responsibility for the originality of the entire content.*

Date: Rajesh T. A.

Dedicated

To my

Mummy (Omana), Pappa (Ayyappen),

Wife (Seena), and Son (Rishabh)

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Rajesh T. A.

Abstract

Aerosols exhibit large spatio-temporal variabilities in their optical, physical and chemical properties, and can influence our planet by interacting with incoming solar and outgoing terrestrial radiation. The objectives of the thesis are to characterize the spatial and temporal variabilities in optical and physical properties of aerosols, source apportion black carbon aerosols, and to estimate the aerosol radiative forcing and their seasonal variability over distinct environments (urban, and high altitude remote). Aerosol characteristics were measured and examined over an urban (characterized by high aerosol concentrations dominated by anthropogenic aerosols) (Ahmedabad (23.03^{o}N, 72.55^{o}E, 55 m above mean sea level (AMSL)), and a high altitude remote region (with low aerosol concentration dominated by transport mechanisms) (Gurushikhar (24.65^{o}N, 72.78^{o}E, 1680 m AMSL). These study locations in western India are influenced by similar meteorology. The influence of atmospheric aerosols on the Earth-atmosphere radiation budget is examined using radiative transfer model. The shortwave aerosol radiative forcing is estimated using two single scattering albedo (*SSA*) values, one derived from the surface measurements of aerosol scattering and absorption coefficients (Method 1), and the other derived from remote sensing satellite measurement (Method 2). Further, to delineate the impact of black carbon (BC) aerosols on the Earth-atmosphere radiation budget the shortwave radiative forcing is computed for BC aerosols only over both the study locations.

Over the urban site, Ahmedabad, high values of scattering (β_{sca}) and absorption (β_{abs}) coefficients are found during morning and late evening due to a substantial increase in the anthropogenic activities and the atmospheric boundary layer dynamics. The scattering and absorption coefficients decrease as day advances (due to the evolution of the atmospheric boundary layer) and

attains a minimum value around afternoon. On the contrary, β_{sca} and β_{abs} over Gurushikhar are higher in the afternoon hrs when compared to forenoon and night because of atmospheric boundary layer dynamics which when accompanied with strong thermal convection aid an upward movement of pollutants to the observational site from the surrounding foothills.

The surface single scattering albedo shows a rare diurnal variability over Gurushikhar when compared to Ahmedabad. The near surface *SSA* is lower over Ahmedabad than Gurushikhar due to the dominance of absorbing aerosols over Ahmedabad from the anthropogenic emissions. The diurnal variation in *Ångström exponent* (α), *backscatter fraction* (*b*), and *asymmetry parameter* (*g*) over Gurushikhar do not show any morning or evening peaks as observed over Ahmedabad consistent with β_{sca} and β_{abs} variations. The maximum α observed during winter suggests the dominance of smaller size aerosols. The minimum α and *b*, and maximum *g* found during monsoon suggest the dominance of larger particles reaching the observational site from the marine region (Arabian Sea). The aerosol optical depth (*AOD*) over Gurushikhar is lower than Ahmedabad, as Ahmedabad is consistently influenced by the high magnitude of anthropogenic emissions, whereas the remote high altitude Gurushikhar is influenced by local and longrange transported aerosols.

The black carbon (BC) mass concentrations, and its equivalent BC from fossil fuel (BC_{FF}) and wood burning (BC_{WB}) exhibit strong diurnal variations over Ahmedabad compared to Gurushikhar due to the combined effects of the diurnal evolution of atmospheric boundary layer and consistent anthropogenic emissions. A distinct BC variation is observed over Gurushikhar with an increase in BC concentration during noontime as seen in β_{abs}. The diurnal contribution of BC_{FF} in total BC dominates throughout the day at both the observational sites. The annual mean contribution of BC_{FF} to total BC mass concentration is 80 and 72% over Ahmedabad and Gurushikhar respectively. This comparison indicates that even a high altitude remote site can have com-

parable fossil fuel contribution due to emissions produced over urban regions.

The study highlights the roles of single scattering albedo and aerosol optical depth (*AOD*) in the aerosol radiative forcing estimate. The differences in the forcing (*ARF*) for composite aerosol following Methods 1 and 2 is attributed to the differences in *SSA* values viz; surface and column. *ARF* estimated using surface *SSA* (lower) (Method 1) is always higher than column *SSA* (higher) (Method 2). The spectral aerosol properties for the black carbon (BC) aerosols exhibit significant variation in the *AOD* for BC aerosols only, but *SSA* and *g* remain the same. The forcing for BC aerosols only over Ahmedabad is higher by a factor of 2-3 than Gurushikhar when *AOD* also varies by the same factor, which confirms the linear dependence of *AOD* on the *ARF*. Over an urban location(Ahmedabad), *TOA* forcing is comparable for Method 1 and BC aerosols only, whereas significant variations are found in *SFC* and *ATM* forcing due to *AOD*. On the contrary, the *TOA* forcing flips sign from +ve to -ve following Method 2 as compared to BC aerosols only. Over a high altitude remote location (Gurushikhar), the forcing values are comparable from both the methods as the *SSA* values are comparable. The *TOA* forcing is always negative as *SSA* is higher over Gurushikhar. The study reveals that over an urban and a high altitude remote locations the BC aerosols alone can contribute in the range of 20 to 60% to the shortwave atmospheric forcing.

It is to be noted that when a high altitude remote site is in the same region as that of an urban aerosol source location, and both theses locations are governed by the same meteorology and atmospheric dynamics, then aerosol measurements over the high altitude region can serve as regional background which is the case here. Results indicate that although Gurushikhar is a high altitude remote site, it is significantly influenced by the local and longrange transported aerosols through convection and advection. The study reveals that Gurushikhar lacks anthropogenic emissions and the aerosol properties over Gurushikhar do not exhibit any significant inter-annual variability, confirming that Gurushikhar is a regional background site for aerosols in western

India. These results can be used as inputs in regional and global climate models for the estimation of climate forcing, to further improve our understanding on the spatio-temporal variability and radiative effects of aerosols over different environments.

Key words: *Atmospheric Aerosols, Scattering Coefficient, Absorption Coefficient, Single Scattering Albedo, Ångström Exponent, Backscatter Fraction, Asymmetry Parameter, Aerosol Optical Depth, Black Carbon, Source Apportionment, Radiative Forcing, Heating Rate, Nephelometer, Aethalometer, Remote Sensing, Optical Properties Model, Radiative Transfer Model, Observations, Urban Region, High Altitude Remote Site.*

Contents

List of Acronyms

ABL	Atmospheric Boundary Layer
AIRS	Atmospheric InfraRed Sounder
AMSL	Above Mean Sea Level
AOD	Aerosol Optical Depth
ATM	Atmosphere
ARF	Aerosol Radiative Forcing
BC	Black carbon
CO_2	Carbon dioxide
DISORT	Discrete Ordinates Radiative Transfer
DU	Dobson Unit
ECMWF	European Centre for Medium-Range Weather Forecast
EOS	Earth Observing System
HEPA	High Efficiency Particulate Air
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
IPCC	Intergovernmental Panel on Climate Change
MODIS	MODerate resolution Imaging Spectroradiometer
NCAR	National Center for Atmospheric Research
NCEP	National Center for Environmental Prediction
OMI	Ozone Monitoring Instrument
OPAC	Optical Properties of Aerosols and Clouds
PPM	Part Per Million
PPT	Part Per Trillion
RH	Relative Humidity
RMS	Root Mean Square
SBDART	Santa Barbara DISORT Atmospheric Radiative Transfer
SF	Scaling Factor
SFC	Surface
SSA	Single Scattering Albedo
TOA	Top Of the Atmosphere
TRMM	Tropical Rainfall Measuring Mission

Notations and Symbols

α	Ångström exponent
β_{ext}	Aerosol extinction coefficient
β_{sca}	Aerosol scattering coefficient
β_{abs}	Aerosol absorption coefficient
β_{bsca}	Aerosol backscattering coefficient
σ	Mass specific attenuation cross section
λ	Wavelength
τ	Total optical depth
$\tau_{Rayleigh}$	Rayleigh scattering optical depth
$\tau_{aerosol}$	Aerosol optical depth
τ_{mol}	Optical depth due to molecular absorption
$\triangle P$	Pressure difference
A	Filter spot area
ARF_{ATM}	Atmospheric forcing
ARF_{SFC}	Surface forcing
ARF_{TOA}	Top of the atmospher forcing
b	Aerosol backscatter fraction
BC_{FF}	Fossil fuel component of *BC*
BC_{WB}	Wood burning component of *BC*
BC(Day)	Day time BC mass concentration
BC(Night)	Night time BC mass concentration
$BC(Day)_{FF}$	Fossil fuel component of *BC(Day)*
$BC(Night)_{FF}$	Fossil fuel component of *BC(Night)*
$BC(Day)_{WB}$	Wood burning component of *BC(Day)*
$BC(Night)_{WB}$	Wood burning component of *BC(Night)*
BC(M)	Morning (05:00-10:55 hrs) BC mass concentration
BC(A)	Afternoon (11:00-15:55 hrs) BC mass concentration
BC(E)	Evening (16:00-20:55 hrs) BC mass concentration
BC(N)	Night (21:00-04:55 hrs) BC mass concentration

$BC(M)_{FF}$	Fossil fuel component of *BC(M)*
$BC(M)_{WB}$	Wood burning component of *BC(M)*
$BC(A)_{FF}$	Fossil fuel component of *BC(A)*
$BC(A)_{WB}$	Wood burning component of *BC(A)*
$BC(E)_{FF}$	Fossil fuel component of *BC(E)*
$BC(E)_{WB}$	Wood burning component of *BC(E)*
$BC(N)_{FF}$	Fossil fuel component of *BC(N)*
$BC(N)_{WB}$	Wood burning component of *BC(N)*
C	Enhancement parameter
C_p	Specific heat capacity
f	Filter loading effect compensation parameter
g	Aerosol asymmetry parameter
I	Direct solar irradiance
I_0	Extraterrestrial solar irradiance
m	Air mass
Q_{ext}	Aerosol extinction efficiency
Q_{sca}	Aerosol scattering efficiency
Q_{abs}	Aerosol absorption efficiency
r^2	Coefficient of determination
R(ATN)	Correction for filter loading effect
V	Volume of air through filter spot

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1. *Black carbon aerosol mass concentration, absorption and single scattering albedo from single and dual spot aethalometers: Radiative implications*
   T.A. Rajesh, and S. Ramachandran
   Journal of Aerosol Science, 119, 77-90, 2018.

2. *Characteristics and source apportionment of black carbon aerosols over an urban site*
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3. *Contribution of aerosol components to aerosol optical depth over a semi-arid location in western India*
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5. *Black carbon aerosols over urban and high altitude remote regions: Characteristics and Radiative implications*
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